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The role of biogeochemical dynamics in the alteration of U solid phases under oxic conditions

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Although *in-situ* and *ex-situ* microbial reduction has been demonstrated to reduce actinide groundwater concentrations in aerobic systems, such biological alterations must be considered temporary treatments unless long-term anoxia can be guaranteed. Under oxidizing conditions, the more mobile higher oxidation states of the actinides such as uranium (U), neptunium (Np), and plutonium (Pu) are the thermodynamically favored species. For example, in U ore deposits in which uraninite (consisting of reduced, tetravalent U as UO_{2+x}) is the parent material, exposure to oxidizing conditions results in alteration to U^{6+} minerals with the U^{6+} -phosphates frequently defining the boundaries of the ore body. While microorganisms are undoubtedly present in such systems, their role in such transformations and the ultimate precipitation of the insoluble phosphate phases is not well understood. Furthermore, the impact of wet-dry cycling on such alterations and the presence of other transuranium actinide elements has not, to our knowledge, been studied to any significant extent.

The purpose of the proposed work is to investigate the role of biogeochemical dynamics in the alteration of U solid phases under oxic conditions to form U^{6+} -phosphate phases. We are investigating the role of important bacterial strains (pure cultures) and consortia of bacterial strains isolated from a DOE field site on the alteration of simple U^{6+} oxide hydrates to U^{6+} phosphates and phosphate solid solutions of U^{6+} and other actinides. To address such transformations in systems that reflect both saturated and vadose zone conditions, systems of interest include those that remain constantly hydrated and those that are exposed to wet-dry cycling. The microorganisms that we are studying include pure cultures of *Bacillus sphaericus* (ATCC 14577), *Desulfovibrio desulfuricans*, and *Geobacter metallireducens*. These strains of microbes were selected to reflect a variety of subsurface conditions including aerobic systems, temporarily anaerobic and/or microaerophilic, and systems with high levels of ionizing radiation. Results with pure cultures will be compared with results obtained using consortia obtained from the FRC site. We are determining transformation pathways between the initial U^{6+} oxide hydrate and the secondary solids that form, as well as rates for these transformations. We are monitoring changes in metal oxidation state, and will provide coordination information about the actinides. Our research links important geochemical and microbiological aspects of this problem, and provides a fundamental basis for predicting the complex and dynamic interplay of such biological treatment strategies.

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